

# Local antiferromagnetic correlations in the iron pnictide superconductors $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{Ca}(\text{Fe},\text{Co})_2\text{As}_2$ as seen via normal-state susceptibility

R. Klingeler, N. Leps, I. Hellmann, A. Popa, U. Stockert, C. Hess, V. Kataev,  
H.-J. Grafe, F. Hammerath, G. Lang, G. Behr, L. Harnagea, S. Singh, and B. Büchner  
*Institute for Solid State Research, IFW Dresden, D-01171 Dresden, Germany*  
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We have studied the interplay of magnetism and superconductivity in  $\text{LaFeAsO}_{1-x}\text{F}_x$  and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ . While antiferromagnetic spin density wave formation is suppressed and superconductivity evolves, all samples show a doping independent strong increase of the normal state susceptibility upon heating which appears a general feature of iron pnictides. The data provide evidence for robust local antiferromagnetic correlations persisting even in the superconducting regime of the phase diagram.

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The appearance of high-temperature superconductivity (SC) in  $\text{LaFeAsO}_{1-x}\text{F}_x$  and related iron pnictides  $\text{REFeAsO}_{1-x}\text{F}_x$  (with RE = Ce, Nd, Sm, Gd) and  $\text{AFe}_2\text{As}_2$  (A = Ca, Sr, Ba) renews strong interest in the complex interplay of magnetism and SC.<sup>1,2,3,4,5,6</sup> Similar to cuprate high- $T_C$  superconductors, iron pnictides exhibit SC in magnetic layers of 3d-atoms, i.e. Fe-atoms, which form a regular square lattice. In the parent material  $\text{LaFeAsO}$ , antiferromagnetic (afm) spin density wave (SDW) order evolves below  $T_N \sim 140$  K and a structural transition from tetragonal to orthorhombic symmetry occurs at  $T_S \sim 160$  K.<sup>7,8,9,10</sup> Like in the cuprates, SC is associated with suppression of magnetic order by electron or hole doping but the undoped parent material is not a Mott-Hubbard insulator but a multi-band metal. In superconducting  $\text{LaFeAsO}_{1-x}\text{F}_x$ , i.e. for  $x \geq 0.05$ , the presence of magnetic order has been ruled out experimentally.<sup>11,12</sup> Introducing additional magnetic moments can induce slow spin fluctuations and even static magnetism in the vicinity of the superconducting phase of  $\text{SmFeAsO}_{1-x}\text{F}_x$ <sup>13</sup> but such fluctuations are clearly absent in the La-based compounds. NMR data show that the spin dynamics in  $\text{LaFeAsO}_{1-x}\text{F}_x$  varies markedly with F doping since afm fluctuations which are clearly visible at  $x = 0.04$  disappear for  $x = 0.11$ .<sup>14</sup> In contrast, our data show that local afm correlations can be present in superconducting  $\text{LaFeAsO}_{1-x}\text{F}_x$  and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  up to the overdoped regime. This conclusion is drawn from the temperature dependence of the normal state susceptibility  $\chi_{\text{norm}}$  in  $\text{LaFeAsO}_{1-x}\text{F}_x$  with  $x \geq 0.05$  as well as in superconducting  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  single crystals. Despite the suppression of the afm order and the occurrence of SC,  $\chi_{\text{norm}}$  is still increasing upon heating and exhibits the same slope as in the undoped case. The robustness and generic character of this behavior at elevated temperatures is remarkable since the ground state qualitatively changes upon doping from a magnetically ordered state to a superconducting state.

Polycrystalline samples of  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  with  $0 \leq x \leq 0.15$  were prepared from pure components as de-

scribed in Ref. 15. Single crystals of  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  ( $0 \leq x \leq 0.125$ ) have been grown in Sn flux as will be described in detail elsewhere.<sup>16</sup> The crystal structure and the composition were investigated by powder X-ray diffraction and wavelength-dispersive X-ray spectroscopy (WDX). From the XRD data impurity concentrations smaller than 3% are inferred. In addition, our samples have been characterised in detail by, e.g., magnetisation, electrical resistivity, NMR, microwave and  $\mu\text{SR}$  studies.<sup>10,11,12,17,18,19</sup> Magnetization measurements have been performed in a MPMS-XL SQUID magnetometer (Quantum Design) in the temperature range 2–350 K in a magnetic field of 1 T and between 2 K and 50 K in 2 mT. Superconductivity was found for  $\text{LaFeAsO}_{1-x}\text{F}_x$  with  $x \geq 0.05$  and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  with  $x \geq 0.045$ . For some compositions, a tiny ferromagnetic contribution was detected which indicates a magnetic impurity phase up to  $\sim 0.01\%$ . In the temperature range under study, the corresponding moment is temperature independent and was subtracted from the susceptibility data. For  $\text{LaFeAsO}$ , measurements up to 800 K imply a temperature hysteresis of the magnetisation above  $\sim 500$  K which we attribute to a starting degradation of the sample. Upon F-doping, the onset of this effect is found already at lower temperatures, i.e. down to 330 K. For NMR experiments, oriented powder of  $\text{LaFeAsO}_{0.9}\text{F}_{0.1}$  was formed by grinding the material, mixing with Sty-cast 1266 epoxy and curing in an external field of 9.2 T<sup>17</sup>, while for  $\text{LaFeAsO}_{0.95}\text{F}_{0.05}$  a powder sample was studied. The NMR measurements have been done in a fixed magnetic field of 7.0494 T.

The temperature dependence of the static susceptibility  $\chi = M/B$  in  $\text{LaFeAsO}$  (Fig. 1) shows clear anomalies around  $\sim 150$  K, similar to data in Refs. [7,20,21]. These anomalies are associated with the structural phase transition to orthorhombic symmetry and to the onset of long range afm ordering, respectively.<sup>10</sup> Both phase transitions are clearly visible in the magnetic specific heat which is proportional to  $\partial(\chi T)/\partial T$  (inset of Fig. 1). The anomaly at  $T_S = 156$  K indicates an enhancement of afm correlations at the structural phase transition which

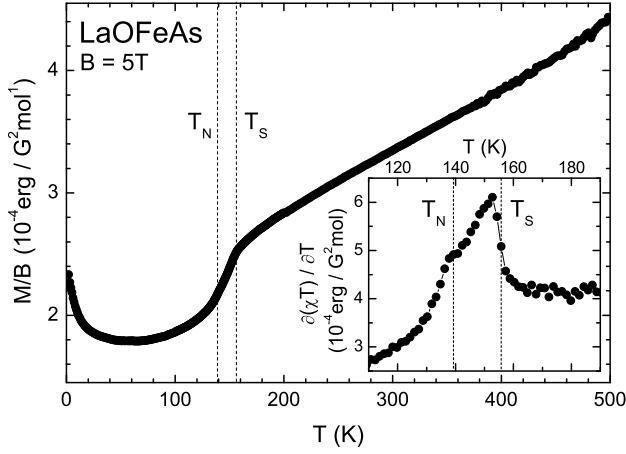


FIG. 1: Static susceptibility  $\chi = M/B$  vs. temperature of LaFeAsO at  $B = 5$  T.  $T_S$  and  $T_N$  label the structural and the afm phase transition, respectively. Inset: Magnetic specific heat  $\partial(\chi T)/\partial T$  in the vicinity of  $T_S$ .

demonstrates an intimate coupling between structure and magnetism. The anomaly of the magnetic specific heat at  $T_N = 138$  K is qualitatively very similar, but much weaker than the one at  $T_S$ . Remarkably, there is a pronounced increase of the susceptibility at higher temperatures, i.e. the susceptibility increases upon heating up to at least 500 K. This strong temperature dependence by nearly a factor of 2 excludes conventional Pauli- and Curie-Weiss-like paramagnetic behavior.

The effect of doping on the magnetic properties of LaFeAsO<sub>1-x</sub>F<sub>x</sub> is presented in Fig. 2. For low doping levels  $x \leq 0.04$  there is only a moderate effect of F-doping on the structural and the magnetic transition. In this low doping regime, the anomalies at the structural and magnetic phase transitions as well as the linear increase of  $\chi$  for  $T > T_S$  are still visible. The susceptibility curves drastically change when the doping level  $x = 0.05$  is achieved where a transition to the superconducting phase is observed at  $T_C \sim 19$  K. In all superconducting samples there is no signature of the anomalies associated to the structural and the magnetic phase transitions. Indeed, the presence of any static magnetic order or structural effects in both underdoped and optimally doped samples was ruled out by recent  $\mu$ SR and thermal expansion studies.<sup>12,22</sup> In addition,  $\chi(T)$  shows no signature of a Curie contribution due to paramagnetic impurity spins, either intrinsic or belonging to a spurious phase. In stark contrast to the strong effect on the ordered phase, the doping effect on the paramagnetic susceptibility is however negligible. For  $x = 0.05$ , e.g., there is a nearly perfect linear  $\chi(T)$  dependence in the temperature regime between  $\sim 30$  K up to at least 380 K. For higher F-concentrations, this behavior is superimposed by a susceptibility upturn in the vicinity of  $T_C$  but there are only negligible changes at room temperature.

In order to prove the intrinsic electronic origin of the experimentally observed static magnetization data

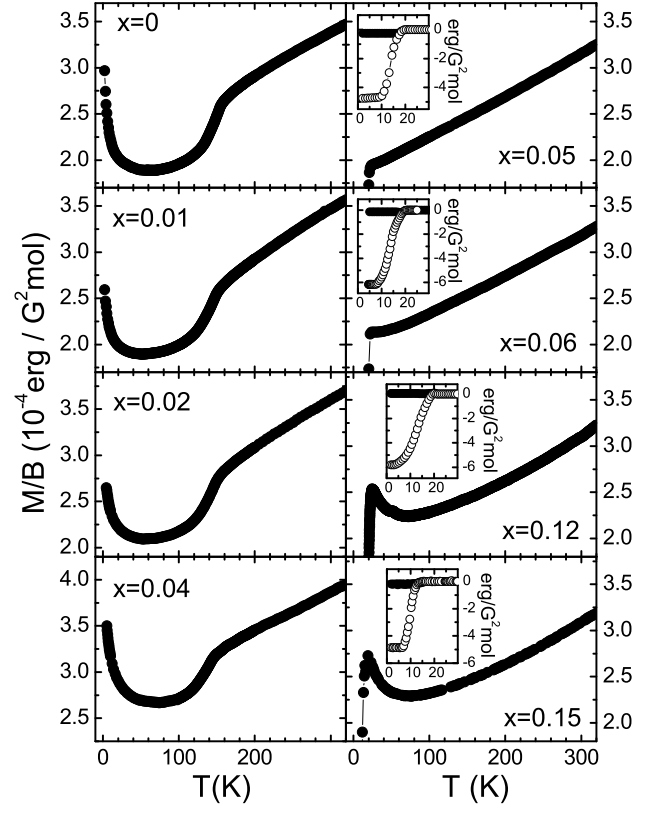


FIG. 2: Static susceptibility  $\chi = M/B$  of LaFeAsO<sub>1-x</sub>F<sub>x</sub>, for different doping level between  $0 \leq x \leq 0.15$  at  $B = 1$  T. Note, that for all graphs the ordinate covers the range  $\Delta\chi = 2 \cdot 10^{-4}$  erg/G<sup>2</sup>mol. Insets:  $M$  vs  $T$  for  $B = 2$  mT.

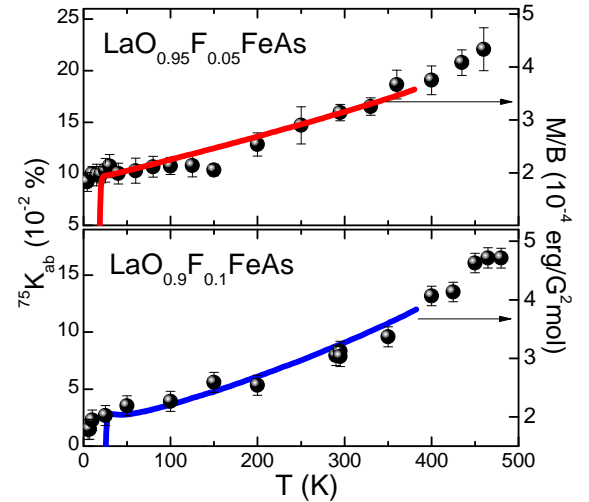


FIG. 3: (Color online) Knight shift  $K_{ab}$  of  $^{75}\text{As}$  (circles) and the macroscopic susceptibility of LaFeAsO<sub>0.95</sub>F<sub>0.05</sub> and LaFeAsO<sub>0.9</sub>F<sub>0.1</sub> versus temperature with different vertical scales and origins.

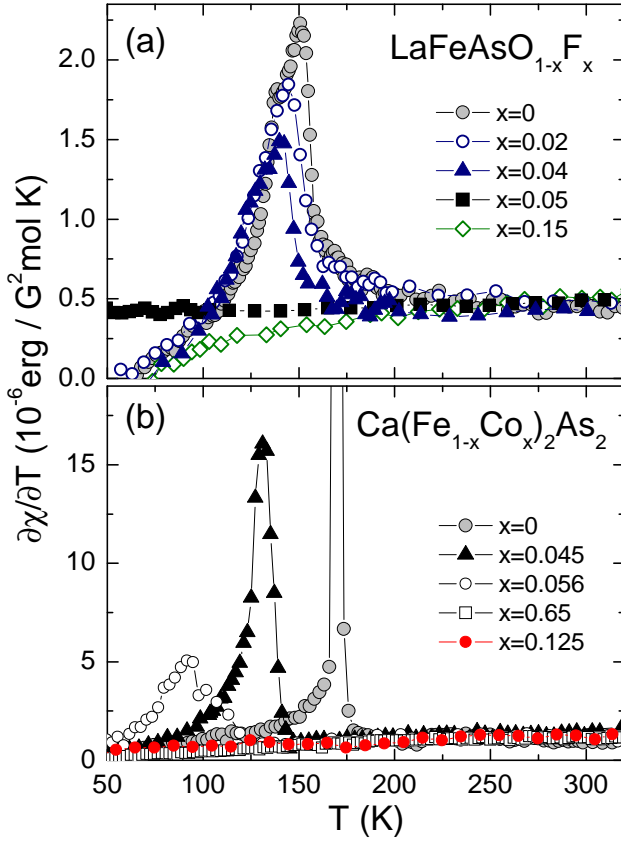


FIG. 4: Derivative of the static susceptibility  $\partial\chi/\partial T$ , at  $B = 1$  T, of  $\text{LaFeAsO}_{1-x}\text{F}_x$  (a) and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  (b), for selected doping levels.

we have investigated the electronic spin susceptibility of the FeAs layers in  $\text{LaFeAsO}_{1-x}\text{F}_x$ , with  $x = 0.05$  and  $x = 0.1$ , as probed by the  $^{75}\text{As}$  nuclear magnetic resonance (NMR) (Fig. 3). Similar to the  $\chi(T)$ -dependence, the in-plane magnetic shift of the  $^{75}\text{As}$  NMR signal  $K_{ab}$  linearly increases from  $T_C$  up to room temperature. We observe a clear scaling of  $K_{ab}$  and macroscopic  $\chi_{\text{norm}}$  as shown in Fig. 3, where the ordinate scales are adjusted to match the curves in the paramagnetic regime. In general, the magnetic shift  $K_{ab}$  consists of the Knight (spin) shift  $K_s \propto \chi_{\text{spin}}$  due to the hyperfine coupling to the electron spins and the orbital shift  $K_{orb}$  which reflects a  $T$ -independent orbital contribution. The fact that the NMR shift scales with the static susceptibility above  $T_C$  implies that the observed macroscopic  $\chi(T)$  is determined by the intrinsic spin susceptibility  $\chi_{\text{spin}}$ . Furthermore, similar to  $K_s$  and  $\chi$  the  $^{75}\text{As}$  longitudinal relaxation rate  $1/T_1$  divided by temperature shows, for  $x = 0.1$ , an increase<sup>17</sup>, roughly yielding a typical for metals constant Korringa product  $S = K_s^2 T_1 T = \text{const}$ <sup>23</sup>. This suggests that  $^{75}\text{As}$  nuclear moments are coupled to itinerant quasiparticles and that itinerant electrons give the main contribution to the static  $\chi$ .

The main result of our susceptibility study is the observation of a similar slope of  $\chi(T)$  in the normal state for

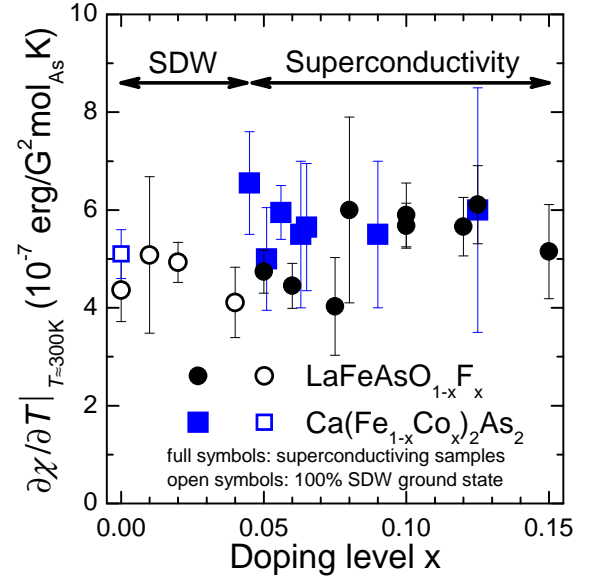


FIG. 5: (Color online) Slope of the susceptibility  $\partial\chi/\partial T$ , at 300 K, as a function of doping in  $\text{LaFeAsO}_{1-x}\text{F}_x$  (circles) and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  (squares). Filled symbols correspond to superconducting samples.

all doping levels under study. This behavior is already evident from the data shown in Fig. 2. A quantitative analysis of the doping effect on the temperature dependence around room temperature is presented in Fig. 5(a), where  $\partial\chi/\partial T$  is shown. For low doping levels  $x \leq 0.4$ , the data exhibit large anomalies which indicate the structural and the SDW transition, respectively. At high temperatures, the data imply  $\partial\chi/\partial T \approx 5 \cdot 10^{-7} \text{ erg}/(\text{G}^2 \text{ mol} \cdot \text{K})$ . Note, that in this low doping regime there are additional contributions to  $\partial\chi/\partial T$  above  $T_S$ , e.g., up to  $\sim 200$  K for  $\text{LaFeAsO}$ , which are associated with concomitant structural and electronic fluctuations.<sup>22</sup> Upon doping, the onset temperatures of these fluctuations as well as  $T_S$  and  $T_N$  are suppressed. In agreement with the complete absence of the orthorhombic and magnetic phases, there are no anomalies in  $\partial\chi/\partial T$  for  $x = 0.05$ , i.e.  $\chi(T) \propto T$ . Here, the slope of  $\chi_{\text{norm}}(T)$  agrees to the one in the tetragonal phase of the low doped compounds. For higher F-concentrations, the additional susceptibility upturn in the vicinity of  $T_C$  yields a temperature regime with  $\partial\chi/\partial T < 5 \cdot 10^{-7} \text{ erg}/(\text{G}^2 \text{ mol} \cdot \text{K})$ , which onset increases upon doping from  $\sim 40$  K at  $x = 0.06$  to  $\sim 175$  K for the highest investigated doping level  $x = 0.15$ . At higher temperatures, however, *all* doping levels  $0 \leq x \leq 0.15$  exhibit the same slope of around  $5 \cdot 10^{-7} \text{ erg}/(\text{G}^2 \text{ mol} \cdot \text{K})$ . This is highlighted by the data in Fig. 5, which shows the slope of the susceptibility  $\partial\chi/\partial T$ , at 300 K, as a function of doping. Within the error bars, we there is no significant dependence on the doping level  $x$ .

Linear temperature dependence of the normal state susceptibility at elevated temperatures seems to be a general feature of iron pnictides. This has been shown, e.g., for undoped 122-materials

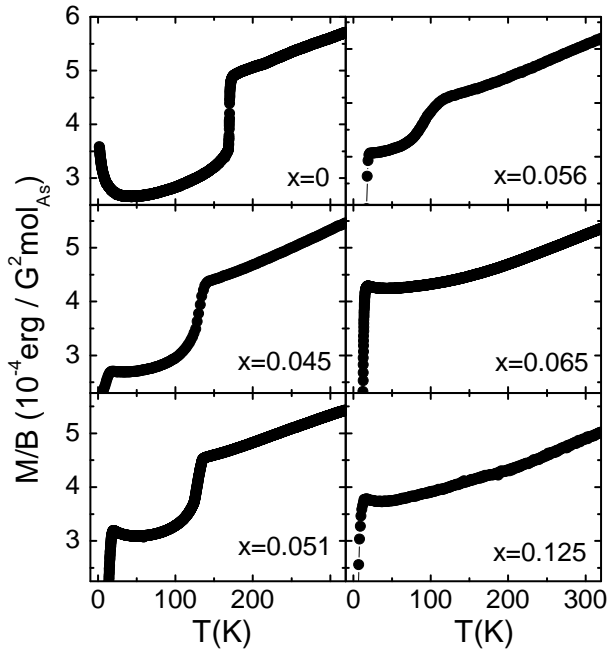


FIG. 6: Static susceptibility  $\chi = M/B$  of  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , for  $0 \leq x \leq 0.125$ , at  $B_{||ab} = 1$  T. For all graphs the ordinate covers the range  $\Delta\chi = 3.5 \cdot 10^{-4}$  erg/G<sup>2</sup>mol.

$\text{AFe}_2\text{As}_2$  ( $A=\text{Ca}, \text{Sr}, \text{Ba}$ ).<sup>24,25,26,27,28</sup> As in the case of  $\text{LaFeAsO}_{1-x}\text{F}_x$ , however, this feature is not restricted to the undoped materials but persists in the superconducting regime of the phase diagram as displayed by the susceptibility data on  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  ( $0 \leq x \leq 0.125$ ) in Fig. 6. For  $x = 0$ , linear behavior is found down to  $T_N = 171$  K, where our crystal exhibits a first-order transition to the SDW-state, which is in a good agreement to Refs. [27,28]. Upon Co-doping, the crystals become superconducting. In addition, anomalies well above  $T_C$  indicate the structural and magnetic phase transitions which temperature at different doping levels is also visible in  $\partial\chi/\partial T$  (Fig. 4(b)). Albeit this qualitative difference compared to  $\text{LaFeAsO}_{1-x}\text{F}_x$ , the paramagnetic susceptibility is very similar and increases linearly with temperature. We note that deviations from linearity as discussed for  $\text{LaFeAsO}_{1-x}\text{F}_x$ , i.e. structural, magnetic and electronic fluctuations above  $T_S$  in the doping regime  $x \leq 0.04$  and superposition of an upturn in the vicinity of  $T_C$  for  $x \geq 0.06$ , are either absent or much less pronounced in  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ . Remarkably, the data not only show a striking qualitative similarity of  $\chi_{\text{norm}}$ , but the slope  $\partial\chi/\partial T$  at 300 K even quantitatively agrees in both materials as shown in Fig. 5. This finding strongly suggests that the increase of  $\chi_{\text{norm}}$  is a robust characteristic property of FeAs-layers in iron pnictides, with both SDW and superconducting ground states and for 1111- as well as 122-materials.

The normal state static magnetic properties in iron pnictides and in particular the slope  $\partial\chi(300\text{ K})/\partial T$  do not significantly depend on the doping level  $x$  or the ac-

tual material under study, i.e.  $\partial\chi(300\text{ K})/\partial T \approx 5 \cdot 10^{-7}$  erg/(G<sup>2</sup>mol<sub>As</sub>K). This holds both for  $\text{LaFeAsO}_{1-x}\text{F}_x$  where the ground state completely changes from an afm poor metal to a non-magnetic SC and for  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  which exhibits an inhomogeneous superconducting state. Neither the suppression of magnetic order and of the structural phase transition nor the evolution of the superconducting ground state strongly affect  $\partial\chi/\partial T$  well above the ordering temperatures. The experimental fact that such a robust feature is present in a large doping range as well as in different materials renders it very unlikely that it depends on subtle details of the Fermi-surface and singular electronic effects. Although we cannot completely exclude any approach critically depending on well-defined nesting conditions, particular band filling or unique band effects, our present data do not affirm such scenarios. This conclusion is corroborated by the absolute size of the observed susceptibility changes. In  $\text{LaFeAsO}$ , the susceptibility changes between 150 K and 500 K amount to  $\Delta\chi \approx 1.9 \cdot 10^{-4}$  erg/(G<sup>2</sup>mol). The electronic density of states from band structure calculations,  $N(E_F)$ , imply a pure Pauli-susceptibility  $\chi_P \sim 5.5 - 7.5 \cdot 10^{-5}$  erg/(G<sup>2</sup>mol) only.<sup>29,30,31,32</sup> Taking into account the moderate mass enhancement of  $\lesssim 2$  (Ref. [7,32]), this is too small to account for the experimentally observed  $\Delta\chi$ , even if changes below 150 K and above 500 K, the robustness of the feature against doping and the size of the susceptibility itself are not considered.

In contrast, we suggest that the magnetic interactions are relevant. The static susceptibility measures the size of magnetization of the material in response to an external magnetic field. Since, as justified above, the measured macroscopic  $\chi$  in the studied iron pnictide samples is the intrinsic spin susceptibility, the experimental data hence imply that the moment per Fe site aligned by the constant external field becomes smaller upon cooling. Such a behavior is typical for the evolution of local afm correlations in systems which are characterized by a spin gap. Typical examples where spin-singlet formation yields  $\partial\chi/\partial T > 0$  are spin ladders, spin dimers, or Haldane chains.<sup>33,34,35</sup> Indeed, recent theories that attempt to describe our data presented above corroborate the scenario of local afm correlations.<sup>37,38,39,40,41,42,43</sup> Even though contradicting approaches are used, starting from the weak or the strong coupling limit, respectively, these studies agree that the experimentally observed increasing  $\chi(T)$  implies the presence of an unusual metallic state with local afm correlations that persist up to high temperature. Zhang et al. apply the 2D frustrated afm Heisenberg model and demonstrate that the data in Fig. 2 provide strong evidence for the existence of a wide afm fluctuation window of *local* magnetic moments.<sup>26</sup> An itinerant approach where nesting boosts  $q = (\pi, \pi)$  SDW magnetic correlations is suggested in Refs. [37,38,39]. In particular, Korshunov et al. show that the linear  $T$ -dependence of  $\chi(T)$  is universal in 2D Fermi-liquids. Remarkably, there is a quantitative agreement between calculated and measured slopes  $\partial\chi/\partial T$  for all doping lev-



els.<sup>39</sup> While this agreement provides evidence for the itinerant approach applied in Ref. 39, the authors identify our experimental data the first observation of a nonanalytic behavior of the 2D spin susceptibility. In contrast, the data in Fig. 2 have been taken as ample evidence for singlet pair formation at elevated temperature in investigations of polaron formation including their dynamics and tendency for binding.<sup>41,44</sup> It is argued that preformed singlet pairs with a binding energy of 100 meV or more would straightforwardly explain the experimental results.

We note, however, that the static susceptibility data alone do not exclude an electronic origin of the unusual temperature dependence  $\chi(T)$ . In addition, a relation of the  $T$ -dependence of the NMR quantities  $K_s$  and  $T_1^{-1}$  to a possible pseudospin gap in the normal state has been controversially discussed in NMR works<sup>14,17,45</sup>. The experimental observation of a nearly constant value  $\partial\chi/\partial T$  would, however, lead to the remarkable conclusion of a doping independent pseudogap in the complete doping range  $0 \leq x \leq 0.15$ . Such a behavior would be completely different to the findings in the superconducting cuprates.<sup>46</sup>

Following the scenario of spin-singlet formation, our experimental results suggest local afm fluctuations in the complete doping regime under study. The doping independent positive slope of  $\chi(T)$  straightforwardly implies that not only the character of these local magnetic fluctuations but also the strength is preserved even if the ground state changes from the afm ordered to the SC

one. In this context we emphasize that  $\text{LaFeAsO}_{1-x}\text{F}_x$  is the only pnictide system which exhibits a homogeneous superconducting phase and the absence of any magnetic and/or structural order in the entire normal state. Thus it is particularly suited to study intrinsic electronic and magnetic properties avoiding complications due to inhomogeneities.

In conclusion, we have studied the static magnetic properties of  $\text{LaFeAsO}_{1-x}\text{F}_x$  and  $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  upon doping. Our data confirm suppression of the structural transition and afm SDW formation and evolution of SC. Our main result is the observation of an increasing paramagnetic susceptibility whose slope is independent of the doping level and the material. Its intrinsic nature is confirmed by measurements of the NMR Knight shift. Our data suggest that strong local afm correlations are present in a broad region of the phase diagrams of iron pnictides and in particular persist in the normal state of superconducting materials.

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